



High-resolution ARPES study of FeSe superconductors

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論文目次

1 Introduction1

1.1 Introduction to Fe-based Superconductors..... 2

1.1.1 Crystal Structure2

1.1.2 Electronic Structure4

1.1.3 Magnetic and Structural Transitions in Fe-based ^[1]_{SEP} Superconductors9

1.1.4 Electronic Nematicity10

1.1.5 In-plane Lattice Strain Effect in Fe-based Superconductors14

1.1.6 High-*T_c* Superconductivity in Atomically Thin Films17

1.2 Research Purpose^[1]_{SEP}22

References23

2 Angle-Resolved Photoemission Spectroscopy28

2.1 Basic Principle of Photoemission Spectroscopy28

2.1.1 Photoemission Spectroscopy28

2.1.2 Angle-Resolved Photoemission Spectroscopy (ARPES)30

2.2 ARPES instrumentation^[1]_{SEP}33

References^[1]_{SEP}37

3 Fabrication of FeSe Thin Films38

^[1]_{SEP}3.1 MBE Instrumentation38

3.2 Fabrication of FeSe Thin Films.....41

3.3 Alkali-Metal Deposition.....42

3.4 Characterization of Pristine FeSe Films by ARPES.....42

3.5 Characterization of Alkali-Metal Deposited FeSe Films by ARPES44

References45

4 Electronic Nematicity in FeSe.....46

^[1]_{SEP}4.1 Background and Goal of Research^[1]_{SEP}.....46

4.2 Experimental Conditions.....47

4.3 Results and Discussion.....47

4.4 Summary52

References53

5 Effects of Strain on the Electronic Structure, Superconductivity, and Nematicity in FeSe	55
5.1 Background and Goal of Research.....	55
5.2 Experimental Conditions.....	56
5.3 Results and Discussion.....	57
5.4 Summary	64
References ^[1] _{SEP}	65
6 Observation of Superconducting Gaps in Se-rich FeSe_{1-x}Te_x	67
6.1 Background and Goal of Research.....	67
6.2 Experimental Conditions.....	68
6.3 Results and Discussions	68
6.4 Summary	72
References.....	73
7 High-T_c Superconductivity in Heavily^[1]_{SEP} Electron Doped Multilayer FeSe Films.....	75
7.1 Background and Goal of Research.....	75
7.2 Experimental Conditions.....	76
7.3 Results and Discussions	77
7.4 Summary	87
References.....	88
8 Summary	90
8.1 Nematicity.....	90
8.2 Carrier Density.....	90
8.3 Electron Pairing.....	91
8.4 Interfacial Effect	91
List of Publications	92
List of Presentations	93
Acknowledgment.....	95

In this thesis, we have reported the evolution of the electronic structure in FeSe by lattice strain, isovalent substitution, and electron doping. Since all these three modifications lead to T_c enhancement, the present results provide important insights into the necessary conditions to realize high- T_c superconductivity in FeSe. In this chapter, we summarize our findings.

In bulk FeSe, we have found evidence for the emergence of nematicity which lifts the d_{xz} and d_{yz} orbitals degeneracy at low temperature. The observed nematicity cannot be explained by the orthorhombic structural transition or long-range antiferromagnetic order. Therefore, the nematicity is likely electronic in origin. We found that this nematicity is suppressed by all of compressive strain, Te substitution, and electron doping. The resultant anticorrelation between the nematicity and T_c suggests that the suppression of nematicity is a necessary condition to realize high- T_c superconductivity.

Besides the nematicity, another intriguing electronic characteristic of bulk FeSe is a low carrier density, as represented by extremely small Fermi surfaces. We have revealed that the compressive strain, Te substitution, and

electron doping commonly increase the summation of hole and electron Fermi-surface volumes, leading to an increase of total carrier density. The observed positive correlation between the total carrier density and T_c indicates an important contribution of large carrier density to T_c enhancement.

The origin of the carrier density enhancement is case dependent. In the case of strained and Te-substituted FeSe, an increase of semimetallic band overlap, which is defined as the energy difference between the hole-band top and electron-band bottom in the normal state, results in an expansion of both hole and electron Fermi surfaces while keeping the carrier compensation condition. Therefore, the semimetallic band overlap may be an important parameter to control physical properties in compensated FeSe. On the other hand, in the electron-doped case, the increase of carrier density is simply caused by a doping-induced expansion of electron Fermi surface.

It is remarked that both the enhanced semimetallic band overlap and the electron doping results in a shift of the van Hove singularity in the normal state toward a higher binding energy. Such a shift would suppress the Pomeranchuk-type instability predicted to stabilize the nematicity, and hence it can be also responsible for the suppression of nematicity.

In Te-substituted FeSe, we have observed a large superconducting-gap size on the electron pocket compared to the hole pocket. It has been also demonstrated that high- T_c superconductivity in electron-doped FeSe originates from the Cooper pairing on the electron pocket. These results suggest that the enhanced pairing on the electron pocket plays an important role for high- T_c superconductivity.

We have estimated the contributions of electron doping and lattice strain to high- T_c superconductivity in FeSe thin film on SrTiO₃. Our estimations suggest that these two effects are not sufficient to explain the high- T_c value in 1 ML film. Therefore, there would be an additional contribution which appears only in the 1 ML film, e.g. interfacial electron-phonon coupling. The T_c enhancement by such effects has been estimated to be ~ 20 K. This implies the existence of a new route to realize high- T_c superconductivity by a material design based on a heterostructure.

鉄系超伝導体 FeSe では、圧力印加、等価数元素置換、キャリア注入、単層膜化など、多様な手段によって超伝導転移温度(T_c)の上昇が報告されており、高温超伝導機構の解明に向けた研究が進められている。本論文では、格子歪み、等価数元素置換、電子ドーピングが FeSe の電子状態に与える影響について角度分解光電子分光(ARPES)を用いて明らかにした。

本論文では、まず FeSe バルク試料($T_c = 8 \text{ K}$)の ARPES 測定を行い、他の鉄系超伝導体と同様に半金属的なバンド構造が実現していることを示した。また、低温でバンド分裂が生じることを明らかにし、そのバンド分裂が回転対称性の破れた電子ネマティック状態の形成に起因すると結論した。

続いて、面内に圧縮歪みを導入した FeSe/CaF₂ 薄膜($T_c = 12 \text{ K}$)、及び等価数元素置換を行った FeSe_{0.8}Te_{0.2}/CaF₂ 薄膜($T_c = 22 \text{ K}$)の ARPES 測定を行った。その結果、 T_c が高いほどホールバンドと電子バンドのエネルギーの重なりが大きくなり、半金属的なバンド構造を保ったままホール及び電子キャリア量がともに増加することを明らかにした。また、 T_c が高いほど低温のバンド分裂が小さいことも見出した。これらの結果から、キャリア量の増大と電子ネマティック状態の抑制が高温超伝導の実現に重要であると結論した。

SrTiO₃ 基板上に分子線エピタキシー法を用いて FeSe 薄膜を作製し、その表面に Li や Cs 原子を吸着して電子をドーブした際の電子状態や T_c の変化についても測定した。その結果、アルカリ金属の種類によらず、十分な電子キャリアをドーブすると低温のバンド分裂が抑制され、それに伴って $T_c \sim 40 \text{ K}$ の高温超伝導が発現することを明らかにした。このことから、ホール及び電子キャリアが補償している場合と同様、電子ネマティック状態の抑制が高温超伝導発現に重要であると結論した。

これまでほとんど未解明であった FeSe バルク単結晶のバンド構造を決定し、さらに格子歪み、等価数元素置換、電子ドーピングによる T_c の上昇とバンド構造の変化の関係を網羅的に明らかにしたこれらの成果は、自立して研究活動を行うに必要な高度の研究能力と学識を有することを示している。したがって、Phan Giao Ngoc 提出の博士論文は、博士(理学)の学位論文として合格と認める。